

RADIONUCLIDE CAPTURE USING MEMBRANES

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Abstract¹

Technology is being developed to provide efficient chemical separations techniques for the analysis and treatment of radionuclide containing waste left behind from the production of nuclear weapons at DOE sites.

The waste varies in composition over extreme ranges of pH from acidic to basic. Bulk liquid and solid extraction processes can be used to remove the various radionuclides prior to vitrification or grouting but suffer from severe limitations.

Discussion

3M's Separation Technology provides the capability of removing contaminants down to detection levels at high flow rates, if necessary in the presence of high levels of radiation. The technology, which may be used for the preparation of samples for analysis, the remediation of contaminated liquids or the retrieval of valuable materials, depends on the ability to make effective separations using very small (10 μ m) particles of adsorbers of various classes. The sorbent is loaded into a web or membrane which is used in a filtration-like process. It

is used as a flat disk^{1,2,3,4} for analytical sample preparation or as cartridge for bulk separations. The prototype of equipment which is scalable to run at 50 gallons per minute using established technology has been developed and tested. The full processing system is projected to have a lower capital cost and smaller footprint than conventional systems.

Technology benefits include:

(i) For Rapid Field Sampling and Analysis:

Increased analytical accuracy and precision together with reduced sample turn-around time, higher productivity and reduced cost per sample.

(ii) For Remediation: A highly efficient ion specific separation system which reduces contaminants to a lower level than that reached on a working basis by ion exchange columns, operating at high flow rates, and concentrating waste in a small volume with the generation of a minimum of secondary waste in a fully engineered system.

Use of the Field Sampler to monitor progress in bulk separation/remediation applications will result in additional gains in productivity.

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Rapid field sampling and analysis

This program, initiated in FY 97, seeks to develop and apply commercial expertise with solid phase extraction membranes to the area of liquid sampling and analysis for radiochemical and heavy metal analytes.

Technetium and lead were chosen as the demonstration analytes. Rugged polypropylene holders for membrane disks were fabricated and demonstrated in a prototype sampling system. Demonstrations were performed at four sites with improvements in the system occurring after each use. Data collected shows comparable results between the disk and baseline methods. (Figure 1)

In addition, on-site analysis techniques using disk color development and other technologies were initiated in laboratory studies. A field transportable beta spectrometer was also demonstrated.

Waste remediation and industrial processing

A number of demonstrations^{5,6,7,8,9} have been conducted at DOE sites ranging in scale from the initial work in a hot cell at INEEL where strontium -90 (⁹⁰Sr) and cesium -137 (¹³⁷Cs) were removed from acidic tank waste on the 100 ml scale to the most recent demonstration at Paducah, using one of the Spiral Wound cartridges discussed below, where technetium -99 (⁹⁹Tc) was removed from 22,000 gallons of groundwater. At Hanford the pilot scale unit was used to remove ¹³⁷Cs and ⁹⁰Sr from ground water and fuel storage water at the 100- N area

Additional, larger scale demonstrations are

planned in order to show that the success achieved in the initial demonstrations may be implemented at a remediation site scale, i.e. so that the technology may be deployed rapidly at DOE sites.

Some highlights of Phase I include:

- The development of a new Spiral Wound adsorber cartridge that has greater capacity and provides more effective separation performance than the previously used pleated cartridges.

Spiral Wound cartridges result in more effective use of the active particle, and therefore fewer cartridges are required to handle a specific radionuclide challenge. This is an advantage, not only in reducing total cartridge cost and the frequency of cartridge replacement but particularly in reducing the overall cost of disposal of fully loaded cartridges, i.e. life cycle cost. E.g. a pleated and a Spiral Wound cartridge were each produced using a WWLTM membrane loaded with Calgon PCB Grade Carbon and challenged with a 10 ppm solution of 2-nitrophenol at a flow rate of one gallon per minute. At 50% breakthrough the pleated cartridge had a 2-nitrophenol capacity of 12.9% per gram of carbon while the Spiral Wound cartridge had a capacity of 27.8%. . We believe that flow patterns in a spiral wound configuration result in more complete utilization of the adsorber membrane as compared with a pleated cartridge, particularly at higher flow rates and when longer cartridges are used.

Pressure drop across a pleated cartridge had the potential of causing pleat collapse or to create a membrane break: the design of the Spiral Wound cartridge removes these concerns, allowing the cartridge to be operated at the maximum pressure which can be tolerated by the other components of the system. (Typically these are rated at 150 psi although as of this time we have not operated a cartridge at a pressure drop of more than 90 psi).

Spiral Wound cartridges have a very simple design making them easier to manufacture consistently: this will be an advantage on commercialization.

The full range of adsorbers which have been incorporated into pleated cartridges can be produced as Spiral Wound cartridges including CoHex, SLP, CST, sodium nonatitanate, SuperLig™ 304, carbon, ATS, C-18 bonded silica. In addition to utilizing 3M WWL™ membranes, Spiral Wound cartridges can be formed from many materials including 3M Empore™ membranes and WWL™ membranes in which the base fiber is polyethylene.

- Progress was made in the development of the next generation cartridge which will have similar performance characteristics to the Spiral Wound cartridge but will require little, if any, prefiltration of challenge solutions.
- Methods were developed for

converting specific adsorbers for cesium and strontium from very fine powders to 10µm particles which can be incorporated into webs and cartridges having a low, (desirable), pressure drop.

- As discussed above a Pilot Scale experiment, demonstrating the ability of Spiral Wound cartridges of WWL membranes loaded with SLP resin to remove technetium was run at the DOE Paducah, KY site.
- Four patent applications were filed.

Advantages over baseline technology

a. Rapid Field Sampling

The end user sampling needs have been identified from site personnel input and incorporated into the overall design and performance specifications. From the results to date, DOE can expect to realize the following benefits based on the successful implementation of the field sampler:

- * Rapid deployment of available technology for characterization purposes with user friendly systems
- * Higher productivity of samples per day over conventional sample preparation procedures (5X)
- * Reduced sample turnaround due to the preparation steps being accomplished at the time of sampling from weeks to days or hours
- * Increased analytical accuracy and precision
- * High rate of incorporation of technology into practice
- * Efficient sampling (>95 % of analyte retained)

- * Selective capture of analyte eliminates potential interferences associated with conventional analytical sample preparation
- * Ability to achieve lower detection levels of analyte by control of sample volumes processed through the membrane
- * Regulatory acceptance
- * Reduced costs in sample handling and analyses
- * Compact, transportable and rugged for field usage

b. Remediation.

All of the demonstrations described in references 5 through 9 involved removal of contaminants from basins and waste streams where the DOE has active remediation programs. The technology developed within this program is capable of reducing the level of contaminants to a lower level than that achieved by ion exchange beds on a daily working basis (e.g. at West Valley and Test Area North at INEEL) and results in a lower level of waste for ultimate disposal. The need for a low volume of waste for ultimate disposal is expected to become increasingly important as more facilities are remediated and the waste generated must be shipped off site. When full life cycle costs for ultimate waste disposal are factored into the economic analysis of the baseline technology we expect that the full benefits of this new technology will be seen.

Technology will be developed in time for deployment by DOE

The program is ahead of schedule for the work proposed for Phase I. We expect limited deployment in the latter part of Phase II as part of a systematic product

introduction.

a. Rapid Field Sampling

The project capitalizes on 3M's proven capability of packaging surface reactive particles of a wide variety of reactive chemistries into porous membranes. By choosing or developing solid phase extraction particles with a high degree of specific selectivity for the target analytes the sample preparation process can be reduced to simply passing the sample fluid through the membrane followed by relatively straight forward analytical methods of detection.

b. Remediation.

The underlying technology has been developed, we are proposing larger scale demonstrations in order to show that the success of the initial demonstrations may be implemented at an industrial scale, i.e. so that the technology may be deployed rapidly at DOE sites. In light of our experience to date^{5,6,7,8,9} we expect to be able to overcome the inevitable difficulties associated with the scale up of any process in a timely way.

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Figure 1

**Comparison of Tc Rad Disk Method
vs Baseline Methods**

